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Implications of the glacial CO_2 "iron hypothesis" for Quaternary climate change

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[1] The "iron hypothesis" posits a role for increased supply of mineral aerosol to the ocean surface during glacial periods in driving atmospheric CO_2 lower; that changes in CO_2 and climate strongly affect dust supply raises the possibility of feedback. Here I take a systems view in analyzing the properties and implications of such a feedback and consider three primary state variables that can be related empirically to each other: dust supply, atmospheric CO_2, and "climate" (surface air temperature). The results of this analysis suggest that the dust-CO_2-climate feedback is primarily an *intra*glacial phenomenon, when it can account for about a third of the temperature variability recorded in Antarctic ice cores. Since glacial-interglacial cyclicity prior to ca. 800 kyr BP is characterized by the absence of a "full" glacial state (such as the Last Glacial Maximum), it is possible that destabilization of climate by the marine iron cycle is fundamental to the differences between "41 kyr" and "100 kyr" climatic regimes. The critical role played by the state of the land surface in this feedback also has implications for the longer-term evolution of the Earth system during the Cenozoic.

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1. Introduction

[2] The influential "iron hypothesis" for glacial-interglacial control of the concentration of CO_2 in the atmosphere [*Martin*, 1990] is based on the premise that biological productivity in the modern Southern Ocean is limited by insufficient supply of the micro-nutrient iron. This posits that limitation is at least partly relieved during glacial times by the enhanced dust deposition to the Southern Ocean suggested by Antarctic ice core records [*Delmonte et al.*, 2002; *Petit et al.*, 1999], the result being enhanced productivity and a draw-down of CO_2. The plausibility of this explanation for lower glacial CO_2 is supported by numerical models of the ocean carbon cycle [*Bopp et al.*, 2003; *Watson et al.*, 2000]. It has also found some support in the results of recent open ocean iron enrichment experiments [*Boyd*, 2002].

[3] In a wider climatic context, taking a "defocused, Earth System" perspective [*Schellnhuber*,

1999], it is clear that the iron hypothesis mechanism is itself part of a ''feedback'' system [*Arimoto*, 2001; *Ridgwell and Watson*, 2002]. This arises because while climate is responsive to dust – through its influence on marine productivity, atmospheric CO_2, and thus the degree of surface warming driven by the ''greenhouse effect,'' dust, in turn, is responsive to climate – the entrainment of dust from the land surface is facilitated by low soil moisture levels and by the absence of vegetation cover [*Harrison et al.*, 2001], while its transport to the open ocean becomes more efficient under a weaker hydrological cycle [*Andersen and Ditlevsen*, 1998; *Yung et al.*, 1996]. Atmospheric CO_2 changes can also have a direct ''fertilizing'' effect on vegetation growth [*Cowling*, 1999], affecting vegetation cover and thus dust entrainment. The ''dust-CO_2-climate'' feedback system encapsulating the ''iron hypothesis'' is outlined schematically in Figure 1. Because the overall sign of the loop is positive, perturbation of any element in the system will be amplified. By having a destabilizing influence on the climate system, the marine iron cycle may play a fundamental role in the substantial climatic variability which characterizes the late Quaternary. The purpose of this study is to elucidate the general properties of the ''dust-CO_2-climate'' feedback as well as its potential role in glacial-interglacial climate change.

2. Methods

[4] The dust-CO_2-climate feedback encapsulating the ''iron hypothesis'' (Figure 1) is a complex and interconnected system, containing multiple subsidiary feedback loops. While not considered here, there are numerous other linkages and loops related to dust. For instance, presence of dust aerosol in the atmosphere has a direct effect on the radiative balance of the planet [*Tegen et al.*, 1996], as well as indirectly via a possible nucleating influence on the cloud formation [*Sassen*, 2002]. There is also a possible feedback on ocean productivity through relationships linking iron-fertilization of the biota, dimethylsulphide (DMS) emissions, and iron solubalization [*Zhuang et al.*, 1992]. Complex systems such as these, often involving poorly quantified biogeochemical transformations and pro-

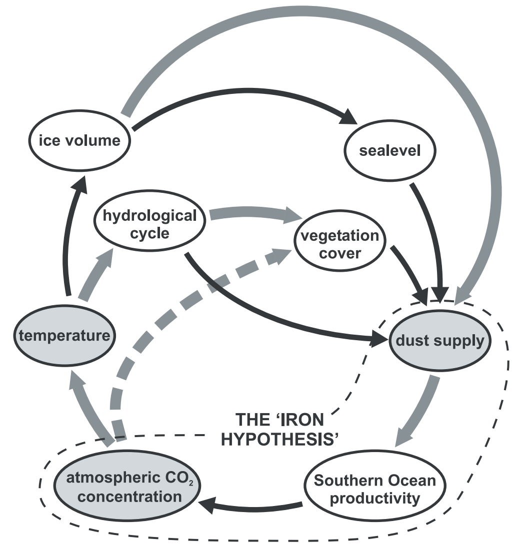

Figure 1. Schematic of the feedbacks in the climate system involving the ''iron hypothesis'' of *Martin* [1990] (shown delineated by a thin dotted line). Different components of the Earth System can be directly related in two possible ways: (i) with a positive correlation (i.e., an increase in the state of one component causes an increase in a second, or, a decrease in the state of one component causes a decrease in a second), shown in gray; or (ii) with a negative correlation (i.e., an increase in the state of one component causes a decrease in a second, or vice versa) shown in black. If a path of successive connections can be traced from any given component back to itself, a closed or ''feedback'' loop is formed. An even number (including zero) of negatively correlated connections counted around the loop gives a positive feedback, which will act to amplify a perturbation of any component within this loop (and tend to destabilize the system). Conversely, an odd number of negative correlations gives a negative feedback overall, which will tend to dampen any perturbation, thus stabilizing the system. Additional interactions peripheral to the arguments presented here have been omitted for clarity. Four main (positive) feedback loops exist in this system, each having a total of either two or four negatively correlated connections within the loop. Working clockwise from dust, these are as follows: (1) dust→productivity→CO_2→ temperature→ hydrological cycle→dust (2 negative correlations), (2) dust→ productivity→CO_2→temperature→hydrological cycle→vegetation→dust (2 negative correlations), (3) dust→productivity→CO_2→temperature→ice volume→dust (2 negative correlations), and (4) dust→ productivity→CO_2→temperature→ice volume→sea level→dust (4 negative correlations). To simplify the feedback analysis, the (positive feedback) loop involving the ''CO_2 fertilization'' influence on the terrestrial biota (shown as a dashed arrow) is omitted.

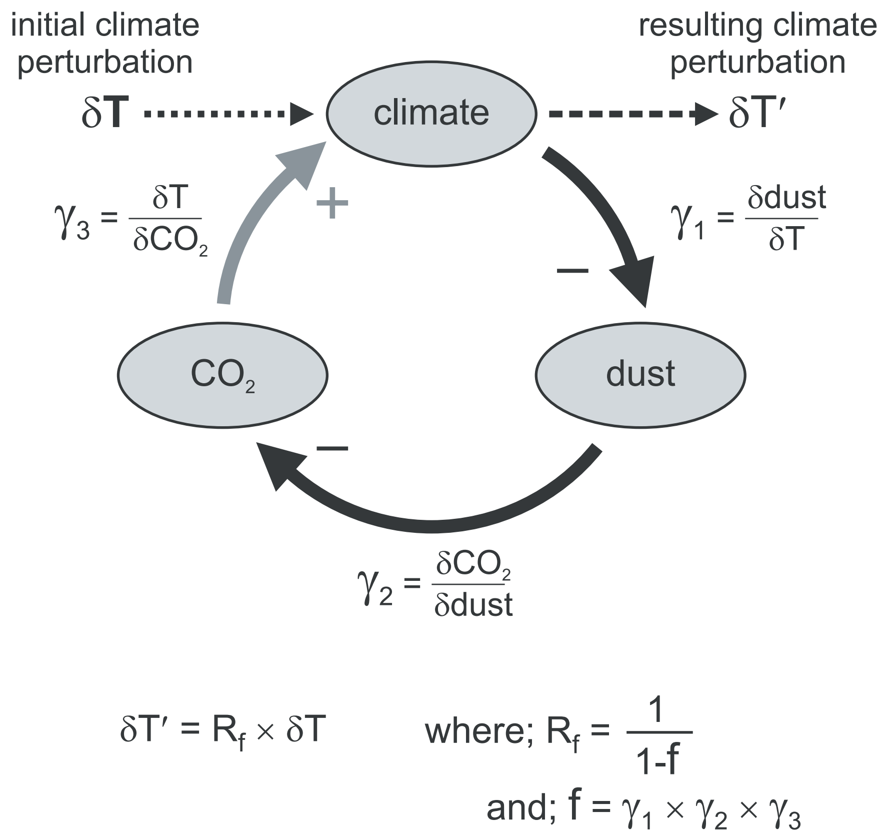

Figure 2. Schematic diagram of the reduced "dust-CO_2-climate" feedback system, comprising the state values of just three primary components (corresponding to the shaded components in Figure 1); dust supply, atmospheric CO_2, and climate (temperature), together the relationships linking them; "dust supply→CO_2," "CO_2→climate," and "climate→dust supply." In this reduced system, two of the component pairs in the loop are negatively correlated and the other positively, giving positive feedback overall (calculated from the product of the signs around the loop). A factor F is defined as the product of the sensitivities (1st derivatives) of the relationships, representing the "feedback" or "system gain." The amplification of a perturbation is then derived. Although the operation of the feedback loop is shown configured with respect to a perturbation of climate (δT), it could equally as well be drawn with respect to a perturbation in either dust supply or atmospheric CO_2. The "iron hypothesis" [*Martin*, 1990] is represented by the arrow: "dust supply→CO_2."

cesses, do not at present readily lend themselves to interrogation by means of mechanistic/processed-based modeling. Thus, although feedbacks are increasingly being recognized in coupled model systems [e.g., *Bendtsen*, 2002; *Claussen et al.*, 2001; *Cox et al.*, 2000; *Friedlingstein et al.*, 2001; *Soden et al.*, 2002], analysis of their role in stabilizing or destabilizing different elements of the Earth System is often restricted to contrasting model behavior with or without the feedback enabled.

[5] A novel analytical approach is taken here. The feedback loop surrounding the "iron hypothesis" is reduced to the state values of just three primary components: dust supply (at high Southern latitudes), the concentration of CO_2 in the atmosphere, and climate, together with the relationships linking them: "dust supply→CO_2," "CO_2→climate," and "climate→dust supply" (shown in Figure 2). To further simplify the analysis, proxies for "dust supply" and "climate" are derived from the Vostok ice core data. The choice of the Vostok dust record is justified because the Antarctic continent is adjacent to the Southern Ocean – the most critical region with respect to changes in iron supply and the potential magnitude of CO_2 drawdown. Dust fluxes are reconstructed from the observed concentration signal [*Petit et al.*, 1999] and estimated snow accumulation history [*Ritz et*

al., 2001; J. R. Petit, personal communication, 2000]. Deep sea sediment core records from the Southern Ocean cannot provide a better proxy for surface flux because the aeolian signal contained in the sediments is heavily overprinted by detrital deposition [*Ridgwell and Watson*, 2002]. The reconstructed (deuterium-excess corrected isotopic) local temperature at Vostok [*Cuffey and Vimeux*, 2001; *Vimeux et al.*, 2003] is used as a proxy for climatic state. The proxy record chosen need not be representative of global climate per se – the requirement of this analysis is that the relationships defining the feedback loop are internally consistent; i.e., if a record of local Antarctic temperature is used to quantify the relationship "climate→dust supply," the climatic effect of a change in CO_2 (CO_2→climate) must also be with respect to Antarctic temperature. The relationship "climate→dust supply" is quantified by regression of dust deposition against temperature (Figure 3a). Because both these variables are contained within the bulk ice, no issues with mismatching chronologies arise.

[6] The relationship "dust supply→CO_2" is quantified with the aid of a carbon cycle model with explicit representation of iron biogeochemical cycling in the ocean [*Ridgwell*, 2001; *Watson et al.*, 2000]. A sensitivity analysis is carried out (Figure 3b), with dust supply to the ocean surface increased from present-day to present-day plus twice the present-day to Last Glacial Maximum (LGM) difference. Although the critical region of interest in the dependence of CO_2 on dust is the Southern Ocean [*Watson et al.*, 2000], dust fluxes are modified over the entire ocean surface to allow like-for-like comparison to be made with the sensitivity exhibited by an alternative model [*Bopp et al.*, 2003]. Omission in the model configuration used here of the response of deep-sea sediments, such as to changes in the calcium carbonate to POC "rain ratio" [*Archer and Maier-Reimer*, 1994] can be justified on similar grounds. However, the disparity between the characteristic time constants of atmospheric CO_2 response to changes in marine iron cycling and sedimentary carbonate preservation, suggests that ocean-sediment interactions are unlikely to play a significant role in determining the strength of the dust-CO_2-climate feedback. While ocean chemistry and atmospheric CO_2 adjusts to a change in "rain ratio" on a timescale of ca. 7 kyr [*Ridgwell et al.*, 2002a], the marine iron inventory and atmospheric CO_2 will respond to dust [*Ridgwell et al.*, 2002b], and climate to CO_2 [*Cubasch et al.*, 2001], both with a timescale of ca. 300 yr.

[7] The results of coupled ocean-atmosphere general circulation models can be used to help characterize the third and final relationship, "CO_2→climate." With respect to year 1990 atmospheric CO_2 concentration of 352 ppmv, equilibrium climate sensitivities (T_{2x}) for a doubling of CO_2 range from about 2.5 to 5.5°C [*Cubasch et al.*, 2001], reflecting the uncertainty in the predictions of current climate models. However, the proxy for "climate" used in the feedback analysis is regional (Vostok) Antarctic cooling. Antarctic continental surface air temperature (SAT) is more sensitive to a change in CO_2 than mean global SAT [*Cubasch et al.*, 2001]. The value of T_{2x} is therefore increased by 25% to reflect this, giving a range for T_{2x}: 3.1–6.9°C. Antarctic SAT for some CO_2 concentration can then be estimated:

$$T = \frac{T_{2x}}{\ln(2)} \cdot \ln\left(\frac{CO_2}{CO_{2(0)}}\right) \tag{1}$$

where $CO_{2(0)}$ = 352 ppmv. Choosing a mid-range value for T_{2x} (5.0°C), a reduction in CO_2 from 280 to 200 ppmv would result in an Antarctic cooling of a little over 2°C, which is in good agreement with the results of *Yoshimori et al.* [2001]. The value of $\delta T/\delta CO_2$ (Figure 2) can then be calculated as an explicit function of CO_2. Although reported equilibrium climate sensitivities include the effect of sea ice feedback on amplifying the temperature sensitivity to a change in CO_2, the important feedback contribution made during glacial times through the response of Northern Hemisphere ice sheets [*Berger and Loutre*, 1997; *Berger et al.*, 1993] is not accounted for. The assumed range of values for $\delta T/\delta CO_2$ and thus strength of the feedback will therefore be an underestimate.

[8] Amplification of an external forcing or internal stochastic variability by the dust-CO_2-climate feedback can be estimated directly from the three

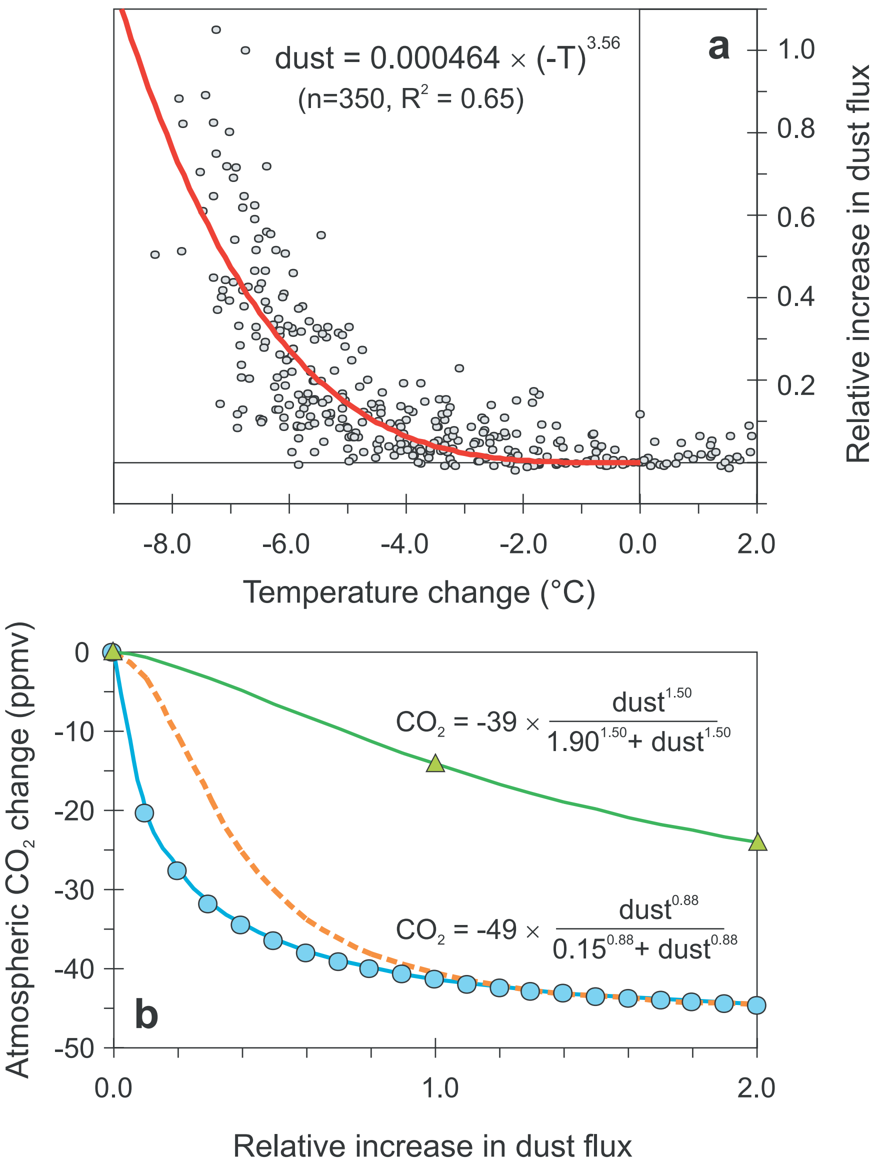

Figure 3. Empirical characterization of the key nonlinear relationships in the feedback loop. a, ''climate→dust supply''; derived from the relationship between dust deposition rates (reconstructed using dust concentration data [*Petit et al.*, 1999] and estimated snow accumulation rates (J. R. Petit, personal communication, 2000)) and isotopic temperature [*Cuffey and Vimeux*, 2001] over the past 350 kyr at Vostok. Data are interpolated at 1 kyr intervals to avoid introducing sampling biasing into the regression. Note that the temperature scale is calculated as a deviation from present-day temperature. b, ''dust supply→CO_2''; derived from model [*Ridgwell et al.*, 2002b] sensitivity analysis of the effect of dust deposition rates on atmospheric CO_2 (blue symbols). Dust flux is scaled so that a value of 0.0 represents a present-day (simulated) deposition [*Mahowald et al.*, 1999] (i.e., no enhancement in dust), and a value of 1.0 represents LGM deposition [*Mahowald et al.*, 1999] (i.e., enhancement by present-day to LGM difference). A value of 2.0 then represents enhancement by twice the present-day to LGM difference. Results of the sensitivity analysis using a 3-D ocean biogeochemical model [*Bopp et al.*, 2003; L. Bopp, personal communication, 2002] are shown in green. A hypothetical curve with the same overall CO_2 sensitivity as the baseline model, but with a different form of response is also shown (dashed orange line).

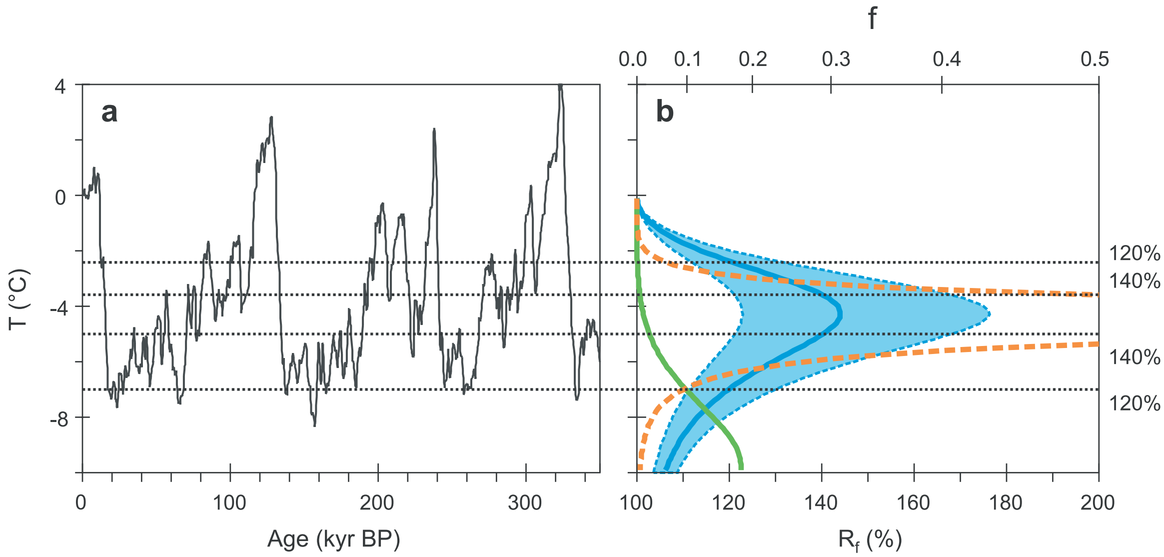

Figure 4. Operation of the "dust-CO_2-climate" feedback. a, Vostok (deuterium-excess corrected) isotopic temperature record [*Cuffey and Vimeux*, 2001]. b, Feedback (f) and net feedback factor (R_f) (equation (3)) plotted as a function of temperature at Vostok. Feedback is calculated with respect to an infinitesimal change in T – the mean feedback across a larger perturbation will be different (since the magnitude of the feedback itself varies with climate state). Predictions using dust-CO_2 sensitivity exhibited by the model of *Ridgwell et al.* [2002b] and mid-range value for T_{2x} (5.0°C) is plotted as a solid blue line, with effect of climate model uncertainty shown as the lighter blue envelope. The Vostok isotopic record (a) is overlain with dashed and dotted lines indicating net feedback factor (R_f) values of 20% and 40%, respectively. Also shown are the predictions arising from use of dust-CO_2 sensitivities exhibited by the model of *Bopp et al.* [2003] (green), as well as the hypothetical dust-CO_2 response curve (orange, dashed) detailed in Figure 3.

relationships which define the loop ("γ_1," "γ_2," and "γ_3" in Figure 2). For an initial temperature perturbation of the system, δT (although it could equally instead be a perturbation of dust or CO_2), if the "feedback factor" of the loop is f, the initial perturbation will be amplified (going around the loop once) to produce an additional temperature change equal to $f \times \delta T$. This will be amplified in turn to produce a further increment, $f \times f \times \delta T$, and so on. The final temperature change ($\delta T'$) is then equal to the initial value (δT) plus the sum of all the additional terms,

$$\delta T' = \left(1 + \sum_i f^i\right) \times \delta T \qquad (2)$$

This can be expressed in terms of the net feedback factor of the system, R_f,

$$\delta T' = R_f \times \delta T; \quad \text{where} \quad R_f = \frac{1}{1-f} \qquad (3)$$

For $0 < f < 1$, $R_f > 1$ and the feedback is positive [*Schlesinger*, 1988], the result being amplification of an initial change in climate (δT). For $f \geq 1$ the sum (equation (2)) does not converge and the net feedback factor of the system is undefined. However, the practical consequences of a situation in which $f \geq 1$ is "runaway" feedback. This is equivalent to the occurrence of a sharp threshold transition in system state.

3. Results

[9] The dust-CO_2-climate feedback (equation (3)) is calculated over the range of observed glacial-interglacial Antarctic climatic (i.e., Vostok temperature) conditions (Figure 4). Amplification via this feedback is not uniform in climate space, but exhibits a pronounced maximum effect centered on a particular intermediate "glacial" state ($T \approx -4.5$ °C). In this respect, it is somewhat akin to the "tuned" amplification of an electrical signal with respect to a particular frequency. This behavior arises because of the antagonistic nature of the two relationships

in the loop, ''climate→dust supply'' and ''dust supply→CO_2.''

[10] At one climatic extreme, as full glacial conditions are approached dust supply to high Southern Hemisphere latitudes becomes increasingly sensitive to small changes in climate and the magnitude of δdust/δT reaches a maximum. This is evident from the form of the highly nonlinear regression of dust flux with temperature(Figure 3a). (As an aside, the high degree of nonlinearity exhibited by this relationship is interesting in itself. It can be understood in terms of the interaction of a number of separate factors, such as decreased vegetation cover and soil moisture content, and greater wind speeds [*Harrison et al.*, 2001; *Tegen et al.*, 1996], and increased efficiency of atmospheric dust transport [*Andersen and Ditlevsen*, 1998; *Yung et al.*, 1996], each individually exhibiting a somewhat lower order dependence on climatic state, but multiplicatively combining to produce the observed high degree of nonlinearity.) However, concurrent with the increasing sensitivity of dust to climate, CO_2 becomes increasingly insensitive to further increases in dust and the magnitude of $\delta CO_2/\delta$dust is small. This is a consequence of the response of biological productivity to changes in iron availability and the onset of limitation by ''secondary'' nutrients such as silicic acid [*Ridgwell and Watson*, 2002; *Watson et al.*, 2000]). Because the ocean iron cycle and biological system ''saturates'' rather more quickly than dust increases, the strength of the resulting feedback weakens as full glacial conditions are approached (Figure 4).

[11] At the other extreme, during interglacials, although the marine iron cycle, biological productivity, and thus atmospheric CO_2 are at their most responsive to changes in dust (and the magnitude of $\delta CO_2/\delta$dust reaches a maximum), dust is relatively unresponsive to climate. Now, the balance in control between the two relationships is reversed, and the insensitivity of dust to changes in climate dominates. This situation also gives rise to weak feedback. Lying between these end-members at intermediate climatic conditions, the strength of the feedback reaches a maximum, with amplification R_f = ~145% (range: 125–175%, taking into account the uncertainty in equilibrium climate sensitivity).

4. Discussions and Conclusion

[12] The analysis presented here suggests that under intraglacial (meaning, mild to full glacial) conditions, operation of the dust supply CO_2-climate feedback can account for ca. one third of recorded Antarctic temperature variability. However, there is an important caveat to this. Because of the central role played by the ''iron hypothesis,'' the results are critically dependent on the predicted response of atmospheric CO_2 to changes in aeolian iron supply, a point on which different models of ocean biogeochemistry currently disagree [*Archer et al.*, 2000; *Bopp et al.*, 2003; *Ridgwell et al.*, 2002b]. This is illustrated by substituting an alternative dust-CO_2 regression for γ_2 (Figure 3b) into the calculation of the loop feedback factor, *F* (Figure 2). Rather than a ''tuned'' response, there is now a continual increase in amplification with cooling climate (Figure 4), and R_f remains <120% over the range of observed Antarctic climatic variability.

[13] There are substantial uncertainties in our current understanding of the ocean carbon/iron cycle and its response to perturbation [*Archer and Johnson*, 2000; *Bopp et al.*, 2003; *Ridgwell et al.*, 2002b; *Wu et al.*, 2001]. Because the marine iron cycle is central to this potential instability (positive feedback) inherent to the climate system, improving the representation of iron biogeochemistry in ocean carbon cycle models may be a prerequisite to gaining a fuller understanding of late Quaternary atmospheric CO_2 and climatic variability. Two specific questions relating to the ocean carbon/iron cycle must be addressed in this respect. The more obvious is ''what is the maximum (glacial-interglacial) response of atmospheric CO_2 to changes in dust deposition?'' For a present-day to LGM increase in dust deposition [*Mahowald et al.*, 1999], current models predict a CO_2 drawdown in the range 5–45 ppm [*Archer et al.*, 2000; *Bopp et al.*, 2003; *Ridgwell*, 2001; *Watson et al.*, 2000]. Models exhibiting lower overall CO_2 sensitivities base their parameterization of iron biogeochemistry on the ubiquitous presence in

the ocean of a iron-binding ''ligand'' [*Archer and Johnson*, 2000; *Archer et al.*, 2000; *Bopp et al.*, 2003; *Lefèvre and Watson*, 1999; *Watson and Lefèvre*, 1999], nominally set at 0.6 nM concentration [*Johnson et al.*, 1997]. This has the effect of buffering the ocean Fe inventory (and thus CO_2) to a dust perturbation. The other end-member approach is to assume no significant role for iron-binding ligands in determining the oceanic distribution of Fe [*Watson et al.*, 2000; *Ridgwell*, 2001, *Ridgwell et al.*, 2002b]. Now, the ocean Fe inventory can ''float'' and the system respond to perturbation, and CO_2 sensitivity is at the higher end of the range. Maybe the ''real'' system lies somewhere in between – although ligands must surely play an important role [*Rue and Bruland*, 1997; *Johnson et al.*, 1997], measured dissolved iron concentrations in the deep Southern Ocean of 0.2–0.3 nM [*de Baar and de Jong*, 2001] are difficult to reconcile with a ligand concentration of 0.6 nM.

[14] The second question is more difficult: ''What is the *form* of the response of atmospheric CO_2 to changes in dust deposition?'' From Figure 3b it is clear that the shape of the relationship ''dust supply→CO_2'' depends critically on how ocean iron biogeochemistry is formulated – the non-ligand model [*Ridgwell et al.*, 2002b] predicts a highly nonlinear response, while the ligand model [*Bopp et al.*, 2003] is not far off linear over glacial-interglacial conditions. This sensitivity to biogeochemical response is further illustrated by considering the implications of a hypothetical curve in which biological productivity is initially relatively unresponsive to increases in dust (Figure 3b). In this case, the value of F exceeds 1.0 over part of the climate span and ''run-away'' feedback occurs. The consequence of this would be the existence of two distinct dust-CO_2-climate states in the Earth System [*Ridgwell and Watson*, 2002] separated by an inherently unstable region.

[15] The lesson here is that it is not only the gross sensitivity (or maximum response) of a particular mechanism that is important to the dynamics of the carbon cycle-climate system, but also the degree of nonlinearity exhibited by that mechanism. This is a property of the system that cannot be diagnosed by standard methodology of making paired comparison between steady state (interglacial) control run and model integration under glacial boundary conditions.

[16] The nature of the dust supply CO_2-climate feedback diagnosed here has implications for earlier Pleistocene and Pliocene time. Apart from a change in characteristic glacial-interglacial periodicity, the transition between 41 kyr (ca. 3.0 to 0.8 Myr BP) and ~100 kyr (0.8 Myr BP to present) ''worlds'' involves a substantial increase in maximum global ice volume [*Raymo*, 1998; *Raymo and Nisancioglu*, 2003]. Because the feedback appears to primarily be an *intra*glacial phenomenon with respect to the 100 kyr cycles, operating at maximum strength between mid- and full-glacial conditions, it is possible that the 41 to 100 kyr transition involves the activation of this feedback – perhaps as a result of a long-term secular trend in boundary conditions [*Paillard*, 1998; *Raymo*, 1997]. This is consistent with the tri-state (''interglacial,'' ''mild glacial,'' and ''full glacial'') conceptual model of *Paillard* [1998, 2001] for the timing of Pleistocene glaciations. If the climate system of the ''41 kyr world'' lacked a sufficiently strong dust-CO_2-climate feedback, the transition to a full glacial state might not be possible, explaining the observed difference in maximum glacial amplitude achieved. While the depth of ''useful'' ice at Vostok limits the core record far short of the transition at ca. 800 ka BP [*Petit et al.*, 1999], the EPICA core drilled on Dome C [*Wolff*, 2002] may provide proxy data reaching far enough back in time to shed new light on the reasons for this change.

[17] The close coupling between global climate and the state of the land surface (particularly vegetation cover) mediated though the ''iron hypothesis'' and changes in atmospheric trace gas composition raises the possibility of analogous positive feedbacks. For instance, if increased prevalence of C_4 plant species on land were to facilitate higher dust fluxes, perhaps in favoring savanna over forest biome types, the following destabilizing positive feedback would be possible: lower CO_2 → spread of C_4 plants → more dust → lower CO_2. The globally synchronous expansion of C_4 grasslands during the late Miocene (7–5 Myr BP) [*Cowling*, 2001] might not then have been a purely

passive response to external (climate and/or atmospheric CO_2) forcing, but could represent the tightly coupled evolution of climate and the terrestrial biosphere.

[18] As the environmental sciences moves toward a more holistic approach to understanding climate change on a range of timescales ("Earth system science"), it is becoming increasingly clear that "feedbacks" are integral to the behavior of the Earth System and its response to both natural and anthropogenic perturbations. However, highly complex and detailed models – increasingly the tools of choice for the analysis of climate change, may not always be the most elucidating. By taking an alternative "defocused" view and utilizing a simple and transparent empirically based model, the potentially central role of the global iron cycle in past climate change is apparent.

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References

Andersen, K. K., and P. D. Ditlevsen, Glacial/interglacial variations of meridional transport and washout of dust: A one-dimensional model, *J. Geophys. Res.*, *103*, 8955–8962, 1998.

Archer, D. E., and K. Johnson, A model of the iron cycle in the ocean, *Global Biogeochem. Cycles*, *14*, 269–279, 2000.

Archer, D., and E. Maier-Reimer, Effect of deep-sea sedimentary calcite preservation on atmospheric CO_2 concentration, *Nature*, *367*, 260–263, 1994.

Archer, D., A. Winguth, D. Lea, and N. Mahowald, What caused the glacial/interglacial atmospheric pCO_2 cycles?, *Rev. Geophys.*, *38*, 159–189, 2000.

Arimoto, R., Eolian dust and climate: Relationships to sources, tropospheric chemistry, transport and deposition, *Earth Sci. Rev.*, *54*, 29–42, 2001.

Bendtsen, J., Climate sensitivity to changes in solar insolation in a simple coupled climate model, *Clim. Dyn.*, *18*, 595–609, 2002.

Berger, A., and M.-F. Loutre, Palaeoclimate sensitivity to CO_2 and insolation, *Ambio*, *26*, 32–37, 1997.

Berger, A., C. Tricot, H. Gallee, and M.-F. Loutre, Water vapour, CO_2 and insolation over the last glacial-interglacial cycles, *Philos. Trans. R. Soc. London, Ser. B*, *341*, 253–261, 1993.

Bopp, L., K. E. Kohfeld, C. Le Quéré, and O. Aumont, Dust impact on marine biota and atmospheric CO_2 during glacial periods, *Paleoceanography*, *18*(2), 1046, doi:10.1029/2002PA000810, 2003.

Boyd, P. W., The role of iron in the biogeochemistry of the Southern Ocean and equatorial Pacific: A comparison of in situ iron enrichments, *Deep Sea Res., Part II*, *49*, 1803–1821, 2002.

Claussen, M., V. Brovkin, and A. Ganopolski, Biogeophysical versus biogeochemical feedbacks of large-scale land cover change, *Geophys. Res. Lett.*, *28*, 1011–1014, 2001.

Cowling, S. A., Simulated effects of low atmospheric CO_2 on structure and composition of North American vegetation at the Last Glacial Maximum, *Global Ecol. Biogeogr.*, *8*, 81–93, 1999.

Cowling, S. A., Plant carbon balance, evolutionary innovation and extinction in land plants, *Global Change Biol.*, *7*, 231–239, 2001.

Cox, P. M., R. A. Betts, C. D. Jones, S. A. Spall, and I. J. Tetterdell, Acceleration of global warming due to carbon-cycle feedbacks in a coupled climate model, *Nature*, *408*, 184–187, 2000.

Cubasch, U., et al., Projections of climate change, in *Climate Change 2001: The Scientific Basis: Contribution of WGI to the Third Assessment Report of the IPCC*, edited by J. T. Houghton et al., pp. 526–582, Cambridge Univ. Press, New York, 2001.

Cuffey, K. M., and F. Vimeux, Covariation of carbon dioxide and temperature from the Vostok ice core after deuterium-excess correction, *Nature*, *412*, 523–527, 2001.

de Baar, H. J. W., and T. M. de Jong, Distributions, sources and sinks of iron in seawater, in *The Biogeochemistry of Iron in Seawater*, edited by D. Turner and R. Hunter, chap. 5, pp. 123–253, John Wiley, Hoboken, N. J., 2001.

Delmonte, B., J. R. Petit, and V. Maggi, Glacial to Holocene implications of the new 27000-year dust record from the EPICA Dome C (East Antarctica) ice core, *Clim. Dyn.*, *18*, 647–660, 2002.

Friedlingstein, P., et al., Positive feedback between future climate change and the carbon cycle, *Geophys. Res. Lett.*, *28*, 1543–1546, 2001.

Harrison, S. P., K. E. Kohfeld, C. Roeland, and T. Claquin, The role of dust in climate today, at the last glacial maximum and in the future, *Earth Sci. Rev.*, *54*, 43–80, 2001.

Johnson, K. S., R. M. Gordon, and K. H. Coale, What controls dissolved iron concentrations in the world ocean?, *Mar. Chem.*, *57*, 137–161, 1997.

Lefèvre, N., and A. J. Watson, Modeling the geochemical cycle of iron in the oceans and its impact on atmospheric CO_2 concentrations, *Global Biogeochem. Cycles*, *13*, 715–736, 1999.

Mahowald, N., et al., Dust sources and deposition during the last glacial maximum and current climate: A comparison of model results with paleodata from ice cores and marine sediments, *J. Geophys. Res.*, *104*, 15,895–15,916, 1999.

Martin, J. H., Glacial-interglacial CO_2 change: The iron hypothesis, *Paleoceanography*, *5*, 1–13, 1990.

Paillard, D., The timing of Pleistocene glaciations from a simple multiple-state climate model, *Nature*, *391*, 378–381, 1998.

Paillard, D., Glacial cycles: Toward a new paradigm, *Rev. Geophys.*, *39*, 325–346, 2001.

Petit, J. R., et al., Climate and atmospheric history of the past 420 000 years from the Vostok Ice Core, Antarctica, *Nature*, *399*, 429–436, 1999.

Raymo, R. E., The timing of major climate terminations, *Paleoceanography*, *12*, 577–585, 1997.

Raymo, R. E., Glacial puzzles, *Science*, *281*, 1467–1468, 1998.

Raymo, M. E., and K. Nisancioglu, The 41 kyr world: Milankovitch's other unsolved mystery, *Paleoceanography*, *18*(1), 1011, doi:10.1029/2002PA000791, 2003.

Ridgwell, A. J., Glacial-interglacial perturbations in the global carbon cycle, Ph.D. thesis, Univ. of East Anglia at Norwich, UK, 2001.

Ridgwell, A. J., and A. Watson, Feedback between aeolian dust, climate, and atmospheric CO_2 in glacial time, *Paleoceanography*, *17*(4), 1059, doi:10.1029/2001PA000729, 2002.

Ridgwell, A. J., A. J. Watson, and D. E. Archer, Modeling the response of the oceanic Si inventory to perturbation, and consequences for atmospheric CO_2, *Global Biogeochem. Cycles*, *16*(4), 1071, doi:10.1029/2002GB001877, 2002a.

Ridgwell, A. J., M. A. Maslin, and A. J. Watson, Reduced effectiveness of terrestrial carbon sequestration due to an antagonistic response of ocean productivity, *Geophys. Res. Lett.*, *29*(6), 1095, doi:10.1029/2001GL014304, 2002b.

Ritz, C., V. Rommelaere, and C. Dumas, Modeling the evolution of Antarctic ice sheet over the last 420,000 years: Implications for altitude changes in the Vostok region, *J. Geophys. Res.*, *106*, 31,943–31,964, 2001.

Rue, E. L., and K. W. Bruland, The role of organic complexation on ambient iron chemistry in the equatorial Pacific Ocean and the response of a mesoscale iron addition experiment, *Limnol. Oceanogr.*, *42*, 901–910, 1997.

Sassen, K., Indirect climate forcing over the western US from Asian dust storms, *Geophys. Res. Lett.*, *29*(10), 1465, doi:10.1029/2001GL014051, 2002.

Schellnhuber, H. J., 'Earth system' analysis and the second Copernican revolution, *Nature*, *402*, C19–C23, 1999.

Schlesinger, M. E., Quantitative analysis of feedbacks in climate model simulations of CO_2-induced warming, in *Physically-Based Modeling and Simulation of Climate and Climatic Change*, edited by M. E. Schlesinger, *NATO ASI Ser., Ser. C*, *243*, 653–736, 1988.

Soden, B. J., R. T. Wetherald, G. L. Stenchikov, and A. Robock, Global cooling after the eruption of Mount Pinatubo: A test of climate feedback by water vapor, *Science*, *296*, 727–730, 2002.

Tegen, I., A. A. Lacis, and I. Fung, The influence on climate forcing of mineral aerosols from disturbed soils, *Nature*, *380*, 419–422, 1996.

Vimeux, F., K. M. Cuffey, and J. Jouzel, New insights into Southern Hemisphere temperature changes from Vostok ice cores using deuterium excess correction, *Earth Planet. Sci. Lett.*, *203*, 829–843, 2003.

Watson, A. J., and N. Lefèvre, The sensitivity of atmospheric CO_2 concentrations to input of iron to the oceans, *Tellus*, *51*, 460–543, 1999.

Watson, A. J., D. C. E. Bakker, A. J. Ridgwell, P. W. Boyd, and C. S. Law, Effect of iron supply on Southern Ocean CO_2 uptake and implications for glacial atmospheric CO_2, *Nature*, *407*, 730–733, 2000.

Wolff, E., Extending the ice core record beyond half a million years, *Eos Trans. AGU*, *83*(45), 509, 2002.

Wu, J., E. Boyle, W. Sunda, and L.-S. Wen, Soluble and colloidal iron in the oligotrophic north Atlantic and North Pacific, *Science*, *293*, 847–849, 2001.

Yoshimori, M., A. J. Weaver, S. J. Marshall, and G. K. C. Clarke, Glacial termination: Sensitivity to orbital and CO_2 forcing in a coupled climate system model, *Clim. Dyn.*, *17*, 571–588, 2001.

Yung, Y. L., T. Lee, C. H. Wang, and Y. T. Shieh, Dust: A diagnostic of the hydrologic cycle during the last glacial maximum, *Science*, *271*, 962–963, 1996.

Zhuang, G., Z. Yi, R. A. Duce, and P. R. Brown, Link between iron and sulphur cycles suggested by detection of Fe(II) in remote marine aerosols, *Nature*, *355*, 537–539, 1992.